Progress in superconductivity Vol.2 No.1 pp 15-19 1 October 2000

Microstructure and Property Relationship of Laser Ablated YBCO Thin Films from Modified Melt-Textured Grown Targets

C. H. Kim*[a], T. S. Hahn[a], K. S. Hong[b]
[a] *Korea Institute of Science and Technology, Seoul, Korea*
[b] *Seoul National University, Seoul, Korea*
Received 10 July 2000

Abstract

$YBa_2Cu_3O_{7-\delta}$ thin films were deposited by laser ablation using modified melt-textured grown targets. As the laser energy density was increased, the films showed an increasing *c*-axis orientation and an improvement of superconducting properties. However, at 4 J/cm^2, the degree of *c*-axis alignment and J_c of the film were considerably reduced. These were attributed to the enhanced *a*-axis outgrowths. It was shown that the increased laser energy density resulted in the formation of Y_2O_3 inclusions during the *c*-axis film growth, and that these inclusions nucleated the *a*-axis outgrowths.

Keywords : Modified melt-textured growth, *a*-axis outgrowth, Y_2O_3 inclusion

I. Introduction

For applications of superconducting thin films to electronic devices such as microwave resonators [1] and digital devices [2], high-quality thin films are prerequisite. To date, $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films are primarily used to fabricate such devices. There has been a number of research dedicated to the deposition of YBCO thin films, however, the optimum range of deposition parameters for preparing high-quality films is quite narrow [3], [4]. Consequently, it is still of significance to understand the microstructure and property relationship of thin films.

Of the various deposition methods, laser ablation is the most popular one [5], [6]. Much effort has been concentrated on the effects of deposition parameters of laser ablation, such as laser energy density [7], substrate temperature [8], and oxygen pressure [9].

In general, laser ablation of YBCO thin films has commonly employed targets fabricated using conventional solid-state sintering. It is known that the surface quality of the target has an influence on the vapor species ejected from the target [10]. Therefore, it is expected that the target would affect the microstructures and properties of the deposited films. In the previous study [11], [12], we employed modified melt-textured growth (MTG) method as well as the conventional sintering to fabricate YBCO targets. Firstly, the effects of the microstructural difference between the two targets were investigated [11]. Secondly, the film microstructures prepared from the two targets were compared [12].

In the present study, we focus on YBCO thin films laser-ablated from the MTG targets. The thin films were deposited at a laser energy density from 1 J/cm^2 to 4 J/cm^2. The microstructures and superconducting properties of the films were investigated, and their relationship was discussed.

*Corresponding author. Fax: +82 2 958 5709
e-mail: chang@kist.re.kr

II. Experimental

The MTG process to fabricate YBCO targets was reported previously [11]: Y_2O_3, $BaCO_3$ and CuO powders were weighed, mixed, and calcined at 880°C for 12 h three times. The calcined powder was pressed into a disk of 1-inch diameter, and then heated up to 1060°C to partially form a liquid phase, quenched down to 1010°C, and finally slow-cooled to 910°C to promote grain growth. The heat-treated disk was annealed in flowing oxygen at 500°C for 10 h to compensate for oxygen loss.

YBCO thin films were deposited on (100) $LaAlO_3$ substrates using a KrF excimer laser (248 nm). While the substrate temperature and oxygen pressure were kept at 760°C and 100 mTorr, respectively, the laser energy density was varied in the range of 1-4 J/cm^2. After the deposition, the vacuum chamber was filled with oxygen up to 500 Torr, and the deposited films were annealed *in situ* at 500°C for 1 h during cool-down. All the films have a thickness of about 200 nm.

Analysis of the phase and orientation for films were performed using an X-ray powder diffractometer (MacScience MPX18, CuK_α). The surface morphology of films was investigated using a field-emission scanning electron microscope (SEM, Hitachi S-4200). The detailed microstructure of films was investigated using a high-resolution transmission electron microscopy (HRTEM, Hitachi H9000-NAR, 300 kV).

To evaluate the superconducting properties, such as critical current (T_c) and critical current density (J_c), of films, the deposited films were patterned into a bridge 400 μm long and about 10 μm wide, and the electrical measurements, such as R-T and I-V, were carried out using the dc 4-probe method. 1 μV criterion was used to determine the critical current.

III. Results and discussion

3.1. Superconducting property

Fig. 1 shows the T_c and J_c (at 77 K) of the films obtained from R-T and I-V measurements. For the laser energy densities in the present study, the films showed T_c of about 84-88 K, and no dependence of T_c on the laser energy density was found. The oxygen

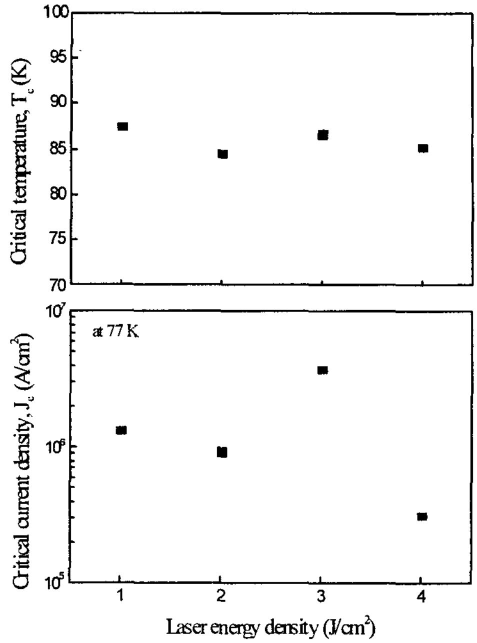


Fig. 1. Critical current (T_c) and critical current density (J_c) of YBCO thin films vs. laser energy density during deposition.

pressures during deposition and in-situ annealing were the same for all of the films such that the oxy gen contents of films were considered to be nearly constant. Unlike T_c, J_c of the films showed some behavior dependent on the laser energy density. As the laser energy density was increased, J_c increased up to the highest value of 3.7×10^6 A/cm^2 at 3 J/cm^2. However, at 4 J/cm^2, the J_c was abruptly decreased. Since J_c is dependent on the film microstructure, the microstructural analyses were carried out to understand the J_c variations.

3.2. Microstructure

Fig. 2 shows the XRD patterns of the films. All of the films were *c*-axis preferred oriented. For comparison, the peaks of the films were normalized with respect to the substrate (200) peak. As shown in Fig. 2, the (00*l*) peaks of the films became stronger and sharper with an increase of the laser energy density, which indicates that the *c*-axis alignment among the grains in the films was improved. At 4 J/cm^2,

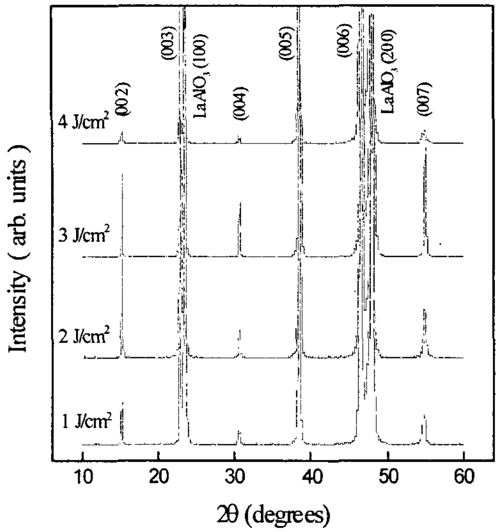
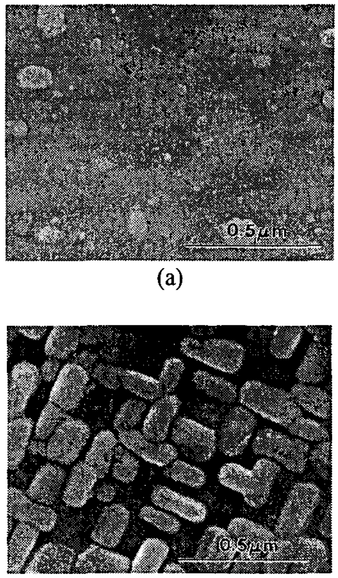


Fig. 2. XRD patterns of YBCO thin films deposited a various laser energy densities. All of the film peaks are normalized with respect to the substrate (200) peak.



(a)

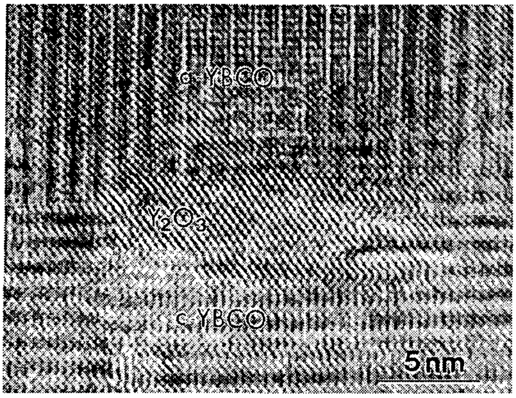
(b)

Fig. 3. Scanning electron micrographs of the surface of the film deposited at (a) 3 J/cm^2 and (b) 4 J/cm^2.

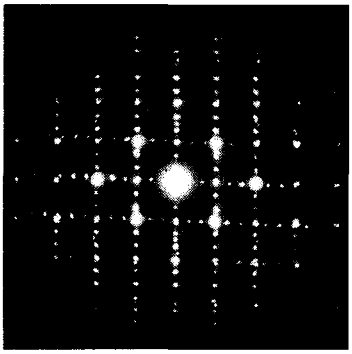
however, the film peaks had considerably reduced intensities with much broader widths. It is considered that this microstructural change is related to the J_c behavior in Fig. 1. To further examine the film microstructures, the surface morphologies of films were investigated.

Scanning electron micrographs of the surfaces of films deposited at 3 J/cm^2 and 4 J/cm^2 are shown in Fig. 3. While the film at 3 J/cm^2 showed rather smooth surface with a few surface outgrowths, the surface of the film at 4 J/cm^2 was remarkably covered with *a*-axis outgrowths [12]. The significantly decreased (00*l*) peaks of the film at 4 J/cm^2, shown in Fig. 2, were attributed to these *a*-axis outgrowths. In addition, the *a*-axis outgrowths would reduce the actual *c*-axis oriented region carrying the supercurrent. This would result in the decrease of J_c. To understand the reason why the *a*-axis outgrowths were considerably formed at 4 J/cm^2, cross-sectional specimen of the film at 4 J/cm^2 was investigated using HRTEM.

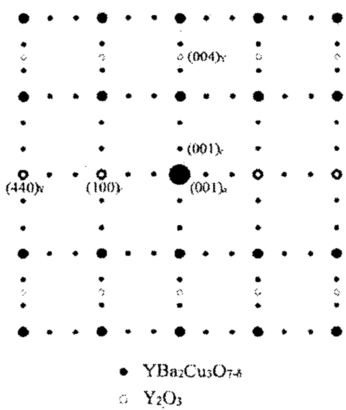
In Fig. 4(a), a high-resolution electron micrograph of the film deposited at 4 J/cm^2 is presented, which shows that *a*-axis grain was heterogeneously nucleated on a second phase during the growth of *c*-axis basal film. Fig 4(b) and (c) show a selected-area electron diffraction pattern and the corresponding schematic for the area in (a), respectively. In addition to the diffraction spots of *c*-axis basal film and *a*-axis outgrowth, those of the second phase appeared. The spots correspond to Y_2O_3 (004) planes, as reported elsewhere [13]. As a consequence, the second phase was identified as Y_2O_3 inclusion with the orientational relationship, $[110]_{Y_2O_3}//[100]_{YBCO}$ and $[001]_{Y_2O_3}//[001]_{YBCO}$ [14]. The nucleation of *a*-axis outgrowth on Y_2O_3 inclusion will be discussed elsewhere [14]. The MTG targets prepared in the present study had a cationic ratio of Y:Ba:Cu=1.25:2:3 [14]. In general, it is known that Y-rich phases on the target surface have a higher ablation threshold [15]. As the laser energy density is increased, the Y-rich phases are easily ablated from the MTG targets. This results in the Y-rich composition of vapor species ejected from the target, and as a result, the formation of Y_2O_3 inclusions in the growing film.



(a)



(b)



(c)

Fig. 4. (a) High-resolution electron micrograph for the cross-section of the film deposited at 4 J/cm^2. *a*-axis grain (denoted as *a*-YBCO) was nucleated on Y_2O_3 inclusion formed in the *c*-axis film (denoted as *c*-YBCO); (b) and (c) selected-area electron diffraction pattern for the area in (a) and the corresponding schematic. The subscripts *a*, *c*, and Y denote *a*-axis and *c*-axis YBCO, and Y_2O_3, respectively.

IV. Conclusion

YBCO thin films were deposited by laser ablation using the targets that were fabricated by modified melt-textured growth rather than conventional sintering. As the laser energy density was increased from 1 J/cm^2 to 3 J/cm^2, the films showed an increasing *c*-axis orientation and an improvement of superconducting properties. However, at 4 J/cm^2, the degree of *c*-axis alignment and J_c of the film were considerably reduced. These were attributed to the enhanced *a*-axis outgrowths, which were nucleated on Y_2O_3 inclusions formed during the *c*-axis film growth.

References

[1] M. R. Rao, "Compact lumped-element microwave resonators using epitaxial $YBa_2Cu_3O_{7-x}/NdAlO_3/$ $YBa_2Cu_3O_{7-x}$ trilayers," Appl. Phys. Lett., 70, 3032-3034 (1997).

[2] W. H. Mallison, S. J. Berkowitz, A. S. Hirahara, M. J. Neal, and K. Char, "A multilayer $YBa_2Cu_3O_x$ Josephson junction process for digital circuit applications," Appl. Phys. Lett., 68, 3808-3810 (1996).

[3] N. G. Chew, S. W. Goodyear, J. A. Edwards, J. S. Satchell, S. E. Blenkinsop, and R. G. Humphreys, "Effect of small changes in composition on the electrical and structural properties of $YBa_2Cu_3O_7$ thin films," Appl. Phys. Lett., 57, 2016-2018 (1990).

[4] R. H. Hammond and R. Bormann, "Correlation between the in situ growth conditions of YBCO thin films and the thermodynamic stability criteria," Physica C, 162-164, 703-704 (1989).

[5] T. Venkatesan, X. Wu, A. Inam, C. C. Chang, M. S. Hegde, and B. Dutta, "Laser processing of high-T_c superconducting thin films," IEEE J. Quantum Electron., 25, 2388-2393 (1989).

[6] B. Roas, L. Schultz, and G. Endres, "Epitaxial growth of $YBa_2Cu_3O_{7-x}$ thin films by a laser evaporation process," Appl. Phys. Lett., 53, 1557-1559 (1988).

[7] T. P. O'Brien, J. F. Lawler, J. G. Lunney, and W. J. Blau, "The effect of laser fluence on the ablation and deposition of $YBa_2Cu_3O_7$," Mater. Sci. Eng., B13, 9-13 (1992).

[8] B. L. Low, S. Y. Xu, C. K. Ong, X. B. Wang, and Z. X. Shen, "Substrate temperature dependence of the texture quality in YBCO thin films fabricated by on-axis pulsed-laser ablation," Supercond. Sci. Technol., 10, 41-46 (1997).

[9] T. Hase, H. Izumi, K. Ohata, K. Suzuki, T. Morishita, and S. Tanaka, "Partial oxygen pressure effects on the

morphology of Y-Ba-Cu-O thin films in laser deposition process," J. Appl. Phys., 68, 374-376 (1990).

[10] V. A. Mal'tsev, Yu. S. Sokolov, A. V. Kulikovsky, M. D. Waterworth, and A. O. Komarov, "Laser fabrication and properties of potassium-doped $Bi_2Sr_2Ca_1Cu_2O_x$ superconducting films," Opt. Laser Technol., 26, 115-117 (1994).

[11] C. H. Kim, I. T. Kim, K. S. Hong, T. S. Hahn, and S. S. Choi, "Effects of target microstructure on pulsed laser deposited $YBa_2Cu_3O_{7-\delta}$ thin films," Thin Solid Films, 358, 223-228 (2000).

[12] C. H. Kim, K. S. Hong, I. T. Kim, T. S. Hahn, and S. S. Choi, "Comparison of microstructures of pulsed laser deposited $YBa_2Cu_3O_{7-\delta}$ thin films using solid-state sintered and modified melt-textured grown targets," Physica C, 325, 127-135 (1999).

[13] P. Lu, Y. Q. Li, J. Zhao, C. S. Chern, B. Gallois, P. Norris, B. Kear, and F. Cosandey, "High density, ultrafine precipitates in $YBa_2Cu_3O_{7-x}$ thin films prepared by plasma-enhanced metalorganic chemical vapor deposition," Appl. Phys. Lett., 60, 1265-1267 (1992).

[14] C. H. Kim, T. S. Hahn, I. T. Kim, and K. S. Hong, "Origin of *a*-axis outgrowth in pulsed laser deposited $YBa_2Cu_3O_{7-\delta}$ thin films from modified melt-textured grown targets," Unpublished.

[15] S. R. Foltyn, R. C. Dye, K. C. Ott, E. Peterson, K. M. Hubbard, W. Hutchinson, R. E. Muenchausen, R. C. Estler, and X. D. Wu, "Target modification in the excimer laser deposition of $YBa_2Cu_3O_{7-x}$ thin films," Appl. Phys. Lett., 59, 594-596 (1991).